Actinides produced by $^{12}C + ^{242}Pu$ and $^{16}O + ^{238}U$ reactions

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The cross sections for ^{250}Fm, $^{244-246}Cf$, $^{242-244}Cm$, and $^{242}Am^{g}$ produced by the $^{12}C+^{242}Pu$ and the $^{16}O+^{238}U$ reactions leading to the same compound nucleus of ^{254}Fm have been measured by using radiochemical methods. The excitation functions show that the difference between the $^{12}C+^{242}Pu$ and the $^{16}O+^{238}U$ reactions can be attributed mainly to the Coulomb barriers. Our results were compared with others reported previously for $^{22}Ne+^{232}Th$, $^{12}C+^{238}U$, $^{12}C+^{240,241}Pu$, $^{13}C+^{241}Pu$, $^{16}O+^{233}U$, and $^{16}O+^{242}Pu$ reaction systems, and support a mechanism involving transfer of α-particle clusters (C,Be,He) from projectile to target for the production of Cf and Cm isotopes.

I. INTRODUCTION

Many actinides have been produced by various heavy-ion-induced nuclear reactions. The reactions, which form actinide isotopes from the targetlike nuclei to the compound nucleus, have been studied recently. For example, the nuclear reactions $^{239}Pu(^{12}C,\alpha 2n\text{-}\alpha 3n)$ and $^{238}U(^{12}C,5n\text{-}6n)$ leading to the same radioactive products ^{245}Cf and ^{244}Cf have been studied by Hahn *et al.*[1] Demin *et al.*[2] have measured excitation functions for production of ^{246}Cf, $^{251,253}Es$, $^{250-254}Fm$, and ^{256}Md in bombardments of ^{249}Cf with ^{22}Ne. Lee *et al.* have investigated the reactions of ^{16}O, ^{18}O, ^{20}Ne, and ^{22}Ne with ^{248}Cm,[3] and ^{18}O with ^{248}Cm and ^{249}Cf.[4] Transfer reaction cross sections of protactinium, uranium, and neptunium isotopes from the interactions of ^{20}Ne and ^{22}Ne with ^{232}Th have been measured by Tanaka *et al.*[5] In the field of heavy actinides, however, it is not easy to investigate such nuclear reactions because of the small reaction cross sections encountered. For the purpose of understanding the nuclear reactions, comparative studies of the different reactions is an effective means in the actinide region. The study of the reactions that occur when the same target is irradiated with several different types of projectiles such as in Ref. 3 is one example. When the kind of projectile ion is fixed and several different types of elements or isotopes are used as targets, other information on the reactions can be obtained. One such study has been reported by Sikkeland *et al.*;[6] enriched uranium isotope targets, ^{233}U, ^{234}U, ^{235}U, and ^{238}U, were each bombarded individually with a ^{12}C ion. We have initiated a systematic study using radiochemical methods to investigate the formation cross sections for actinides produced in bombardment of ^{242}Pu with ^{12}C ions and ^{238}U with ^{16}O ions at energies near the Coulomb barriers.[7] Both reactions form the same compound nucleus, ^{254}Fm, by a complete fusion reaction:

$$^{12}C+^{242}Pu \rightarrow ^{254}Fm^{*} ,$$

$$^{16}O+^{238}U \rightarrow ^{254}Fm^{*} .$$

The investigation of the production of identical actinide isotopes in reactions where the same compound nucleus is formed by different projectile-target combinations is another method which can be used in addition to studies of fixed-projectile and -target reactions. This paper deals with the excitation functions of actinides synthesized in these heavy-ion bombardments and a comparative study of the reactions between $^{242}Pu(^{12}C,4n)^{250}Fm$ and $^{238}U(^{16}O,4n)^{250}Fm$ and between $^{242}Pu(^{12}C,\alpha xn)^{250-x}Cf$ and $^{238}U(^{16}O,\alpha xn)^{250-x}Cf$ $(x=4,5,6)$ as well as the transfer reactions for these systems which lead to $^{242-244}Cm$ and $^{242}Am^{g}$ isotopes. We also report on a comparison of our results with other reaction systems.

II. EXPERIMENTAL

The plutonium (99.8% ^{242}Pu) and uranium (99.98% ^{238}U) targets were prepared by electrodeposition from isopropyl alcohol solution onto aluminum foils of 7 μm thickness. The uranium targets varied in thickness from 0.3 to 2.0 mg/cm^2 and the plutonium target was 0.25 mg/cm^2. A target assembly consisted of a stack of aluminum foils for degrading the beam energy at the upstream side of the target and an aluminum foil (7 μm thickness) for catching the recoil nuclei at the downstream side. Ir-

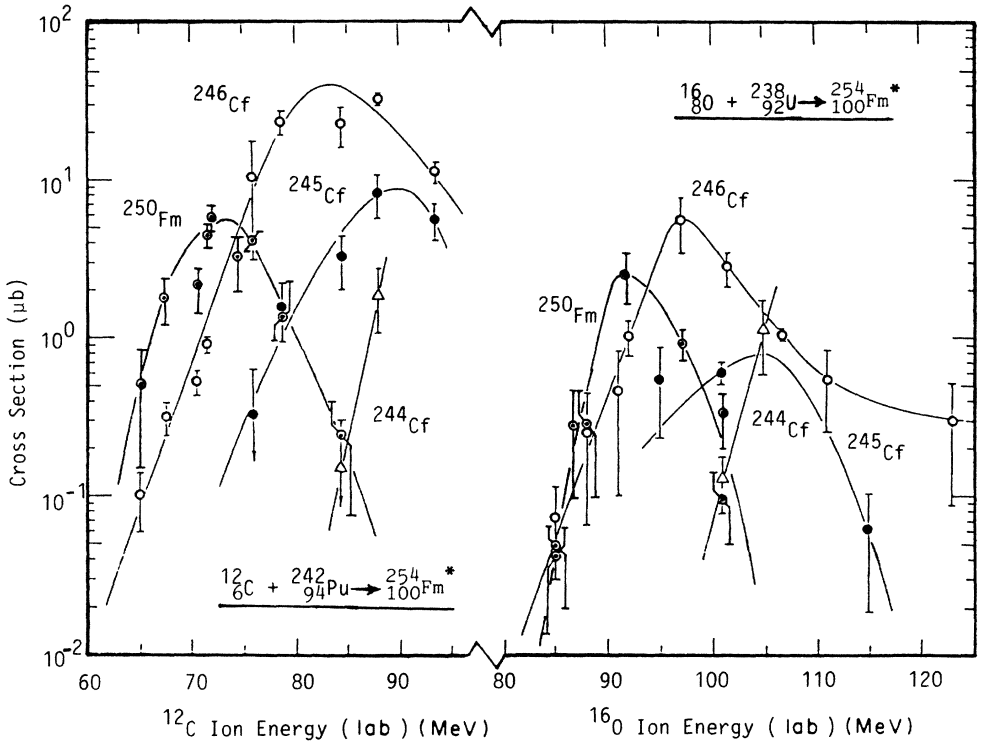

FIG. 1. Excitation functions of ^{250}Fm, ^{244}Cf, ^{245}Cf, and ^{246}Cf produced by the ^{12}C + ^{242}Pu (left side of figure) and the ^{16}O + ^{238}U (right side) reactions.

radiations with ^{12}C and ^{16}O ions were performed at the tandem accelerator of the Japan Atomic Energy Research Institute. The beam energy was varied from 60 to 95 MeV for ^{12}C ions and from 85 to 130 MeV for ^{16}O ions. The beam intensities were up to 200 particle nA for the ^{12}C + ^{242}Pu system and 150 particle nA for the ^{16}O + ^{238}U system, and typical irradiation durations were 0.5 to 3 h. After irradiation, both the target and the aluminum catcher foil were dissolved in aqua regia and the solution was heated to near dryness. The residue was then redissolved in a mixture of 1M nitric acid and 90% methyl alcohol. Each actinide was isolated by elution from a 2 mm diam×80 mm column of MCI GEL CA08Y anion exchange resin (23.5±4 μm) with heated (80–90°C) 0.1–1M HNO_3-80–95% methyl alcohol solutions which were supplied to the column by 1×10^6 Pa pressure.[8] The time for complete separation is about 20 minutes including the source preparation for alpha spectrometry. The chemical yield was determined to be 90±10%. The alpha activities from fermium, californium, curium, and americium isotopes were assayed with Si(Au) surface-barrier detector systems. The measurements were continued for about two months after irradiation. Each actinide nuclide synthesized from the reactions was identified by its chemical behavior, alpha energy, and half-life. The ^{243}Cm and ^{244}Cm isotopes could not be identified separately, because these isotopes have similar alpha energies and half-lives. Detector efficiencies and geometries were determined from measurements of LMRI (Laboratoire de Métrologie des Rayonnements Ionisants, France) standard sources. The cross sections were calculated from the measured activities, the number of ^{242}Pu or ^{238}U atoms in the target, and the integrated beam intensities. It was assumed that all of the actinide products were caught in the aluminum catcher foil. Corrections were made for growth and decay in the case of the parent-daughter isotopes such as ^{250}Fm-^{246}Cf and ^{246}Cf-^{242}Cm. The statistical standard deviations of the counting data and uncertainty of the chemical yield were considered in the calculation of the reaction cross sections. Excitation functions for the actinides produced by the reactions and for the total fission cross section were also calculated with the code ALICE,[9] which can be used to calculate the cross sections of evaporation residues and fission following formation of the compound nuclei through a statistical model.

III. RESULTS AND DISCUSSIONS

The experimental excitation functions for ^{250}Fm produced by the ^{12}C + ^{242}Pu and the ^{16}O + ^{238}U reactions are shown in Fig. 1. The solid lines connect the measured values. The yield curves appear to be symmetric in both reactions. The cross section dependence on the projectile energy in the ^{16}O + ^{238}U reaction differs a little from those reported by others.[10,11] The formation cross section of ^{250}Fm from the ^{12}C + ^{242}Pu reaction is several times larger than that from the ^{16}O + ^{238}U reaction. The difference in the Coulomb barriers between the two reactions is calculated to be 18 MeV, and it is observed from Fig. 1 that the maximum of the excitation function for the ^{16}O + ^{238}U reaction occurs at a projectile energy about 20 MeV higher (18 MeV in the center of mass system) than

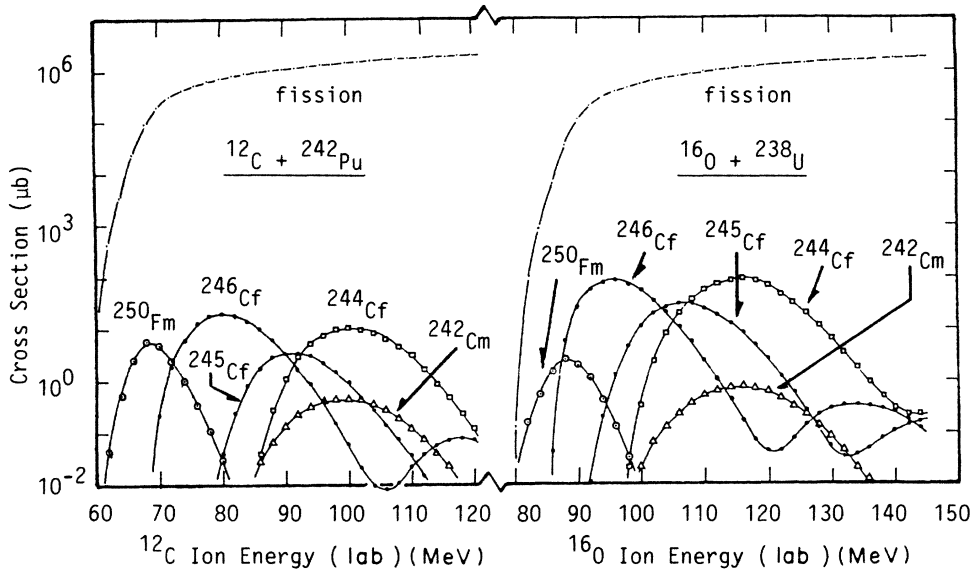

FIG. 2. Calculated excitation functions of ^{250}Fm, $^{244-246}Cf$, ^{242}Cm, and fission in the $^{12}C + ^{242}Pu$ (left side of figure) and $^{16}O + ^{238}U$ (right side) reactions with the code ALICE.

for the $^{12}C + ^{242}Pu$ reaction. The excitation functions calculated with the code ALICE are shown in Fig. 2. The theoretical curves were calculated assuming that the corresponding fission-barrier heights are fixed at 2.00 and 2.31 times the rotating liquid-drop values for all angular momenta,[12] B_f, for the $^{12}C + ^{242}Pu$ and for the $^{16}O + ^{238}U$ systems, respectively, and that the level-density parameters of fission to particle emission ratio, a_f/a_n, are equal to 1.00 for both systems, in order to fit the calculated values to the experimental ones. The calculated excitation functions for ^{250}Fm give the same shapes as the experimental ones showing that the ^{250}Fm isotope is produced via the compound nucleus reactions, $^{242}Pu(^{12}C,4n)^{250}Fm$ or $^{238}U(^{16}O,4n)^{250}Fm$. The fermium isotopes are formed from other projectile-target combination systems: $_{10}Ne + _{90}Th$, $_9F + _{91}Pa$, $_7N + _{93}Np$, $_5B + _{95}Am$, etc. A comparison of the reaction systems which form fermium, californium, and nobelium isotopes by the reaction of neutron evaporation from the compound nucleus is given in Table I. In the comparison between the reactions which form the same compound nucleus of $^{254}Fm^*$ ($^{22}Ne + ^{232}Th$, $^{16}O + ^{238}U$, $^{13}C + ^{241}Pu$, and $^{12}C + ^{242}Pu$), it is observed from Table I that the maximum formation cross sections of the fermium isotopes increase with increasing Z in the target, namely, with decreasing Z in the projectile. Concerning the fixed-projectile reactions ($^{16}O + ^{233}U$ and $^{16}O + ^{238}U$, and $^{12}C + ^{240}Pu$ and $^{12}C + ^{242}Pu$), the cross sections of fermium increase with increasing number of neutrons in the target. These tendencies cannot be explained by only $\Gamma_n/(\Gamma_n+\Gamma_f)$ values which have been calculated by Simbel *et al.*[18,19] In the cases of the fixed-target reactions ($^{12}C + ^{238}U$ and $^{16}O + ^{238}U$, and $^{12}C + ^{242}Pu$ and $^{16}O + ^{242}Pu$), the maximum cross sections formed by the evaporation of four neutrons are determined to be 62 and 1.5 μb for C + U and O + U systems, and 6 and 0.034 μb for C + Pu and O + Pu systems, respectively. These results can be explained from the differences of $\Gamma_n/(\Gamma_n+\Gamma_f)$; the values for C-projectile systems are apparently larger than those for O-projectile systems.

TABLE I. Comparison of the maximum formation cross sections of Fm and Cf isotopes formed by the reaction of evaporation of four neutrons.

Reaction	σ_{max} (μb)	E_p[a] (MeV)	$\Gamma_n/(\Gamma_n+\Gamma_f)$[b]	Ref.
$^{232}Th(^{22}Ne,4n)^{250}Fm$	0.25	107	0.16	13
$^{233}U(^{16}O,4n)^{245}Fm$	0.20	93	0.049	14
$^{238}U(^{16}O,4n)^{250}Fm$	1.5	92	0.093	This work
$^{238}U(^{18}O,4n)^{252}Fm$	0.75	94	0.081	15
$^{240}Pu(^{12}C,4n)^{248}Fm$	1.3	71	0.09	14
$^{241}Pu(^{13}C,4n)^{250}Fm$	5	71	0.11	16
$^{242}Pu(^{12}C,4n)^{250}Fm$	6	72	0.12	This work
$^{238}U(^{12}C,4n)^{246}Cf$	62	67.5	0.28[c]	6
$^{242}Pu(^{16}O,4n)^{254}No$	0.034	89	0.044	17

[a]Projectile energy in lab system at σ_{max}.
[b]References 18 and 19.
[c]Reference 6.

The formation of californium isotopes, ^{244}Cf, ^{245}Cf, and ^{246}Cf, was observed in both reaction systems, and the experimental excitation functions are also shown in Fig. 1. It is seen from the figure that the shapes of the experimental cross sections for ^{244}Cf, ^{245}Cf, and ^{246}Cf isotopes are very similar in both the $^{12}C + ^{242}Pu$ and the $^{16}O + ^{238}U$ reactions, and maxima in the experimental excitation functions for the californium isotopes are shifted by the difference in the Coulomb barriers between the $^{12}C + ^{242}Pu$ and the $^{16}O + ^{238}U$ reaction systems as observed in the case of the fermium isotopes. The calculated formation cross sections for the californium isotopes by the code ALICE, are given in Fig. 2. Table II gives the experimental and calculated maximum formation cross sections of Cf isotopes formed by the reactions of $^{16}O + ^{238}U$, $^{12}C + ^{239}Pu$, and $^{12}C + ^{242}Pu$. Comparing our experimental values with the ones calculated with the code ALICE, the observed cross sections for the production of ^{246}Cf and ^{245}Cf are 6 and 0.8 μb from the $^{16}O + ^{238}U$ system versus 40 and 8 μb from the $^{12}C + ^{242}Pu$ system, respectively, while the calculated cross sections are greater for the O + U (84 and 31 μb) than for the C + Pu (21 and 3.5 μb) system. When our results are compared with any other reaction system, $^{12}C + ^{239}Pu$,[1] the maximum cross section of ^{245}Cf for the $^{12}C + ^{242}Pu$ (8 μb) system is the same as for the $^{12}C + ^{239}Pu$ (7 μb) and greater than for the $^{16}O + ^{238}U$ (0.8 μb) system. For these contradictions there is a possible explanation. The formation cross sections of the californium isotopes should not be calculated with the code ALICE because the code assumes particle evaporation following the fusion reaction. Hahn *et al.*[1] have pointed out that for the $^{12}C + ^{239}Pu$ system an aggregate (a Be fragment) is transferred from the projectile to the target nucleus, followed by evaporation of neutrons from the resulting heavy nucleus. If the Cf isotopes for the O + U system are also produced by the transfer reaction, a C fragment is transferred to the target:

$$_6C + {}_{94}Pu \rightarrow {}_{98}Cf^* + ({}_6C - {}_4Be) ,$$
$$_8O + {}_{92}U \rightarrow {}_{98}Cf^* + ({}_8O - {}_6C) ,$$
$$_{98}Cf^* \rightarrow Cf + xn .$$

Because a Be transfer might be expected to be much more probable than a C transfer, the cross section of Cf isotopes for the C + Pu system probably becomes greater than those for the O + U system. The maximum yield as a function of mass for Cf isotopes from the ^{242}Pu system appears to be at mass 246 or higher, while for the ^{239}Pu system the yields for masses 244 and 245 are nearly the same. This reflects the neutron excess of the target as observed by Lee *et al.*[3]

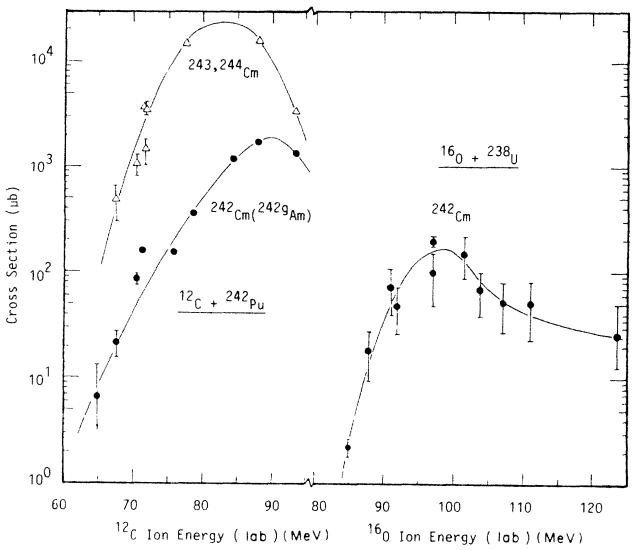

FIG. 3. Excitation functions of ^{242}Cm (^{242}Am^g) and ^{243,244}Cm produced by the $^{12}C + ^{242}Pu$ (left side of figure) and the $^{16}O + ^{238}U$ (right side) reactions.

The isotopes of ^{242}Cm and ^{243,244}Cm were observed in the curium fraction after the chemical separation in the $^{12}C + ^{242}Pu$ reaction system. The ^{242}Cm was also detected in the americium fraction; this means that ^{242}Am^g was produced by the reaction and then decayed to ^{242}Cm. The ratio of the formation cross section of the ^{242}Am^g to that of ^{242}Cm was 3.50 when the ^{12}C ion energy was 78 MeV (lab system). The excitation functions of ^{242}Cm(^{242}Am^g) and ^{243,244}Cm are shown in Fig. 3. In the $^{16}O + ^{238}U$ reaction, only the ^{242}Cm isotope was detected, and the excitation function is also given in Fig. 3. The formation cross sections of the curium isotopes are much larger than those of the fermium and californium isotopes in both reaction systems and apparently differ from the values calculated with the code ALICE (Fig. 2). Sikkeland *et al.*[20] computed the excitation function for the production of ^{242}Cm in the ^{16}O bombardments of ^{238}U using a kinematic model and concluded that the ^{242}Cm was produced by transfer of a Be fragment. Not only ^{242}Cm but also ^{243,244}Cm is probably produced in the present reaction systems by transfer of Be and He fragments to ^{238}U and

TABLE II. Comparison of the maximum formation cross sections of Cf isotopes formed by the reactions of $^{16}O + ^{238}U$, $^{12}C + ^{239}Pu$, and $^{12}C + ^{242}Pu$.

Reaction	σ_{max} (μb) Expt.	σ_{max} (μb) Calc.	E_p[a] (MeV) Expt.	E_p[a] (MeV) Calc.	Ref.
$^{238}U(^{16}O,\alpha 4n)^{246}Cf$	6	84	97	96	This work
$^{242}Pu(^{12}C,\alpha 4n)^{246}Cf$	40	21	83	80	This work
$^{238}U(^{16}O,\alpha 5n)^{245}Cf$	0.8	31	105	106	This work
$^{242}Pu(^{12}C,\alpha 5n)^{245}Cf$	8	3.5	90	92	This work
$^{239}Pu(^{12}C,\alpha 2n)^{245}Cf$	7	10	68	72	1
$^{239}Pu(^{12}C,\alpha 3n)^{244}Cf$	5	9	74	78	1

[a]Projectile energy in lab system at σ_{max}.

^{242}Pu, respectively. This explains the much lower yields for the ^{238}U system since Be transfers have much lower cross sections than He transfers.

In summary, the difference between the excitation functions for ^{250}Fm and $^{244-246}Cf$ produced in the $^{12}C + ^{242}Pu$ and the $^{16}O + ^{238}U$ reactions appears to be explained by the difference in the Coulomb-barrier energies for the two systems. The fermium isotope is produced by neutron evaporation following a fusion reaction, while the californium isotopes are formed by transfers of C fragments for O + U system and Be fragments for the C + Pu system. It is found that the isotopes of $^{242-244}Cm$ are also produced in the $^{12}C + ^{242}Pu$ reaction and ^{242}Cm in the $^{16}O + ^{238}U$ reaction. These isotopes are formed by transfer of Be and He fragments from projectile to target.

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